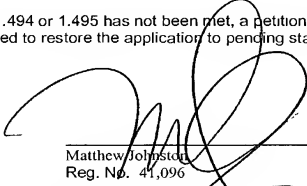


JC20 Rec'd PCT/PTO 20 MAR 2002

FORM PTO-1390 (REV 5-93)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY DOCKET NO. 08292 045	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				DATE March 20, 2002	
				U.S. APPLN. NO. (IF KNOWN, SEE 37 C.F.R. 1.5) Unknown <b>10/088642</b>	
INTERNATIONAL APPLICATION NO. PCT/JP00/06466			INTERNATIONAL FILING DATE September 21, 2000		PRIORITY DATE CLAIMED September 22, 1999
TITLE OF INVENTION. ROLLER FOR ROLLING ELECTRODE STRUCTURE AND THE ELECTRODE STRUCTURE					
APPLICANT(S) FOR DO/EO/US Takaya SATO and Tatsuo SHIMIZU					
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371 (THE BASIC FILING FEE IS ATTACHED)</p> <p>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures [35 U.S.C. 371(f)] at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</p> <p>4. <input checked="" type="checkbox"/> A proper demand for International Preliminary Amendment was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed [35 U.S.C. 371(c)(2)]</p> <p>a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau)</p> <p>b. <input type="checkbox"/> has been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English [35 U.S.C. 371(c)(2)].</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 [35 U.S.C. 371(c)(3)]</p> <p>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 [35 U.S.C. 371(c)(3)].</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) [35 U.S.C. 371(c)(4)].</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 [35 U.S.C. 371(c)(5)].</p> <p>Items 11 - 16 below concern other document(s) or information included:</p> <p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98.</p> <p>12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input checked="" type="checkbox"/> Other items or information: <input type="checkbox"/> Payment by Check attached hereto. <input checked="" type="checkbox"/> Drawings (15 sheets (Figs. 1-15B))</p>					

JC13 Rec'd PCT/PTO 20 MAR 2002

U.S. APPL. NO. (IF KNOWN) SEE 37 C.F.R. 1.57 Unknown <b>107/088642</b>		INTERNATIONAL APPLICATION NO. PCT/JP00/06466		ATTORNEY DOCKET NO. 08292 045 DATE: March 20, 2002	
17. <input checked="" type="checkbox"/> The following fees are submitted: <b>Basic National Fee [37 C.F.R. 1.492(a)(1)-(5)]:</b> Search Report has been prepared by the EPO or JPO.....\$890.00 International preliminary examination fee paid to USPTO (37 C.F.R. 1.482).....\$710.00 No international preliminary examination fee paid to USPTO (37 C.F.R. 1.482) but international search fee paid to USPTO [37 C.F.R. 1.445(a)(2)].....\$1040.00 Neither international preliminary examination fee (37 C.F.R. 1.482) or international search fee [37 C.F.R. 1.445(a)(2)] paid to USPTO.....\$1040.00 International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4).....\$ 100.00				CALCULATIONS   PTO USE ONLY	
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				\$ 890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date [37 C.F.R. 1.492(e)]				\$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	12 - 20 =	0	X \$ 18.00	\$ 0	
Independent Claims	4 - 3 =	1	X \$ 84.00	\$ 84.00	
Multiple dependent claim(s) (if applicable)			+ \$280.00	\$ 0	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 974.00	
Reduction by one-half for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 C.F.R. 1.9, 1.27, 1.28).				\$	
<b>SUBTOTAL =</b>				\$ 974.00	
Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date [37 C.F.R. 1.492(f)].				\$	
<b>TOTAL NATIONAL FEE =</b>				\$ 974.00	
Fee for recording the enclosed assignment [37 C.F.R. 1.21(h)]. The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. 3.28, 3.31). \$40.00 per property				\$ 40.00	
<b>TOTAL FEES ENCLOSED =</b>				\$ 1014.00	
				Amount to be refunded	\$ 0
				Charged	\$ 1014.00
a. <input checked="" type="checkbox"/> A check in the amount of \$1014.00 to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. 50-0548 in the amount of \$ to cover the above fee. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 50-0548.					
NOTE: Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive [37 C.F.R. 1.137(a) or (b)] must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO: Liniak, Berenato, Longacre & White 6550 Rock Spring Drive Suite 240 Bethesda, Maryland 20817 Tel: (301) 896-0600 Fax: (301) 896-0607					
				 Matthew Holyston Reg. No. 41,096	

**PATENT APPLICATION**

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:

Takaya SATO et al.

International Appln. No.: PCT/JP00/06466

Filed: March 19, 2002

Attorney Dkt. No.: 08292.045

For: ROLLER FOR ROLLING ELECTRODE STRUCTURE AND THE ELECTRODE  
STRUCTURE

**PRELIMINARY AMENDMENT**

Commissioner for Patents  
Washington, D.C. 20231

March 20, 2002

Sir :

Prior to the prosecution of the above-captioned application, please enter the following amendments.

**IN THE SPECIFICATION:**

Please amend the Specification as follows:

Please amend the first full paragraph on Page 3 between lines 2-7 as follows:

This invention still is a roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising one work roll and the other work roll having an electrode structure inbetween, a backup roll generating a pressing force onto the surface of the other work roll, a pressuring device pressing the backup roll toward the work roll side, and a drive unit rotating the work roll.

Please amend the fourth paragraph on Page 10 between lines 12-17 as follows:

For example, the adjustment plate 46 may be a spring and a spacer, which maintains clearances among four rolls when a pressuring device 44 such as a hydraulic cylinder is extended. The adjustment plate positioned between the work rolls 43, 43 adjust a gap appropriately so that the work rolls 43, 43 may not contact each other. The spring pushes the

housings 442, 443 and rolls 42, 43 evenly with gaps when the pressuring device such as a cylinder moves backward.

Please amend the fifth paragraph on Page 10, line 19 as follows.

(h) Roller for one backup roll and a pair of work rolls.

Please delete the 7<sup>th</sup> paragraph of Page 13 between lines 20-25.

**IN THE CLAIMS:**

Please amend claim 5 as follows:

5. (Amended) A roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising one work roll and the other work roll having an electrode structure inbetween, a backup roll generating a pressing force onto the surface of the other work roll, and a pressure device pressing the backup roll toward the work roll side, and a drive unit rotating the work roll.

**REMARKS**

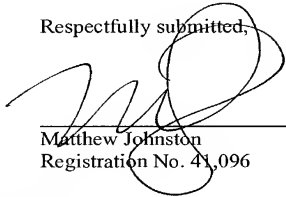
Claims 1-12 are pending in the application. By this Amendment, the Specification and Claim 5 is amended to incorporate the Article 34 Amendment made in the corresponding International Application.

The attached page is captioned "Version with markings to show changes made".

No new matter has been introduced.

Applicants believe that no fee is required for this submission. However, should a fee be due, please charge such fee to Deposit Account No. 50-0548.

Respectfully submitted,



Matthew Johnston  
Registration No. 41,096

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**PATENT APPLICATION**

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:

Takaya SATO et al.

International Appln. No.: PCT/JP00/06466

Filed: March 19, 2002

Attorney Dkt. No.: 08292.045

For: ROLLER FOR ROLLING ELECTRODE STRUCTURE AND THE ELECTRODE  
STRUCTURE

**VERSION SHOWING CHANGES MADE**

Commissioner for Patents  
Washington, D.C. 20231

March 20, 2002

Sir :

Prior to the prosecution of the above-captioned application, please enter the following amendments.

**IN THE SPECIFICATION:**

Please amend the Specification as follows:

Please amend the first full paragraph on Page 3 between lines 2-7 as follows:

This invention still is a roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising [a large diameter work roll and a small diameter other work roll] one work roll and the other work roll having an electrode structure inbetween, a backup roll generating a pressing force onto the surface of [the small diameter] the other work roll, a pressuring device pressing the backup roll toward the [small diameter] work roll side, and a drive unit rotating the work roll.

Please amend the fourth paragraph on Page 10 between lines 12-17 as follows:

For example, the adjustment plate 46 may be a spring and a spacer, which maintains clearances among four rolls when a pressuring device 44 such as a hydraulic cylinder is extended. The adjustment plate positioned between the work rolls 43, 43 adjust a gap





**IN THE CLAIMS:**

Please amend claim 5 as follows:

5. (Amended) A roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising

[a large diameter work roll and a small diameter other work roll] one work roll and the other work roll having an electrode structure inbetween,

a backup roll generating a pressing force onto the surface of [the small diameter] the other work roll, and

a pressure device pressing the backup roll toward the [small diameter] work roll side, and a drive unit rotating the work roll.

12/p.k

## Roller for Rolling Electrode Structure and the Electrode Structure

### Field of the Invention

This invention relates to a coating of an ion-conducting polymer on an electrode structure for an electrical component such as a primary and secondary cell, a capacitor, and an electric double layer capacitor.

### Background of the Invention

In the prior art, in a lithium ion battery, a positive electrode has an electrode structure o is manufactured by mixing a mixture e of a powdered electrode active substance a comprising  $\text{LiCoO}_2$ , powdered electrically-conducting carbon b, a binder polymer c and a solvent d into a slurry, and applying it to a current-collecting member f and drying to form a compound film g, as shown in Fig. 13. The compound film g of Fig. 13 is a partial enlargement of the mixture e on the current-collecting member f.

A negative electrode l as an electrode structure is manufactured by mixing the mixture e of the powdered electrode active substance a comprising powdered graphite, the binder polymer c and the solvent d into a slurry, applying it to the current-collecting member f, and drying to form the compound film g, as shown in Fig. 14.

The electrode structure is designed such that the electrode structure o is sandwiched between two work rolls m, m as shown in Fig. 16; a roller for rolling is used to increase the filling density of the electrode active substance. However, in rolling the electrode structure o is such that the electrode structure o inserted between the upper and lower work rolls m, m is pressed by being sandwiched between the upper and lower work rolls with a pressure force generated by a pressure device n, the weight of the upper work roll, and the weight of the housing p.

In that case, although there is no issue to be resolved if such force is lower, if the

force is higher both upper and lower work rolls becomes distorted. In order to withstand against the distortion, the upper and lower work rolls m, m may be designed thicker. Yet, the radial length pressing the electrode structure o viewing from the sectional direction of the rolls becomes longer, and there is no substantial change in the unit-area pressing force comparing to when the work rolls are in the previous thickness even if the work rolls are thicker.

As shown in Fig. 15, since the conventional lithium ion secondary cell itself has a liquid electrolyte j filled between the positive electrode h and the negative electrode l, appropriate void l is to be remained in the electrode active substance layer (mixture film) flowing the electrolyte j around the electrode active substance, thereby practically functioning as a cell.

However, if a solid electrolyte instead of a liquid electrolyte is used, the solid electrolyte just like the liquid electrolyte cannot flow around the electrode active substance. Therefore, no sufficient current density is attained, which makes it impossible to practically function as a cell.

### **Disclosure of the Invention**

It is an object of this invention to provide a roller suitable for rolling the electrode structure.

It is another object of this invention to provide an effective electrode structure.

(This invention relates to the following the roller.)

This invention is a roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising a pair of work rolls sandwiching and rolling the electrode structure, at least a pair of backup rolls generating a pressing force onto the respective work roll, a pressuring device pressing at least one of the backup roll toward the work roll side,

and a drive unit rotating the work rolls.

This invention still is a roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising a large diameter work roll and a small diameter other work roll having an electrode structure inbetween, a backup roll generating a pressing force onto the surface of the small diameter work roll, a pressuring device pressing the backup roll toward the small diameter work roll side, and a drive unit rotating the work roll.

This invention still is a roller processing/rolling an electrode structure, wherein the roller comprises a work roll housing having an axle receive of the work roll inside and a housing having the respective roll axle inside, and a spacer is provided between the respective two housings.

This invention still is an electrode structure for an electric part where ions move between the electrodes, wherein a powdered electrode active substance coated by an ion-conducting polymer is adhered to a current collecting member, and a roller processes/rolls the powdered electrode conductive substance on the current collecting member.

This invention still further is an electrode structure for an electric part where ions move between the electrodes, wherein a powdered large surface material coated by an ion-conducting polymer is adhered to a current collecting member, and a roller processes/rolls the powdered electrode conductive substance on the current collecting member.

This invention yet further is an electrode structure mixing a powdered electrode active substance with a powdered electrode conducting substance.

### **Brief Description of the Drawings**

Fig. 1 is a diagram showing the manufacture of an electrode structure

comprising an electrode active substance which supplies ions.

Fig. 2 is a diagram of an electrode structure comprising an electrically conducting substance wherein electricity moves between ions.

Fig. 3(A) is a schematic diagram of the roller, and Fig. 3(B) is a diagram showing an electrode structure to be rolled.

Fig. 4 is an isometric view of the roller mounting a pair of backup rollers.

Fig. 5 is an isometric view of the roller mounting one roller.

Fig. 6 is a diagram showing the roller mounting a pair of backup rollers.

Fig. 7 is a diagram showing the roller mounting one backup roller.

Fig. 8 is a schematic view of a secondary cell.

Fig. 9 is a front view of the press-sliding mixer.

Fig. 10 is a front view of the press-sliding mixer.

Fig. 11 is a side view of the press-sliding mixer as shown in Fig. 10.

Fig. 12 is a diagram showing the conventional manufacture of an electrode structure comprising an electrode active substance which supplies ions.

Fig. 13 is a diagram of a conventional electrode structure comprising an electrically conducting substance wherein electricity moves between ions.

Fig. 14 is a schematic view of the conventional secondary cell.

Fig. 15 is a front view of a conventional roller.

This invention will now be described in more detail referring to the drawings.

#### (a) Electrical components

Electrical components wherein ions migrate between electrodes are those in which an ion-conducting substance is disposed between electrode structures

functioning as electrodes, and ions migrate within the ion-conducting substance so that a current flows between the electrodes, e.g., primary cells, secondary cells, and an electric double layer capacitor.

Batteries are characterized in that an ion-conducting substance disposed between the positive electrode structure and negative electrode structure and that an ion, including a proton or a positive hydrogen ion, migrates between electrodes and is accumulated. An electric double layer capacitor is characterized in that an ion-conducting substance is disposed between a pair of electrode structures and an electric double layer is formed between a large surface material within the electrode structures and an electrolyte of the ion-conducting substance.

#### (b) Electrode structures

Electrode structures are used as electrodes in these electrical components, and exchange electrical charges with ions or draw ions. Fig. 13 and Fig. 14 show conventional structures that the powdered electrode active substance a is simply combined with the binder polymer c to adhere to the current-collecting member whereas Fig. 1 and Fig. 2 show the structure that the powdered electrode active substance 11 coated with the ion-conducting polymer 12 is adhered to the current-collecting member 13. For instance, they have a construction in which the powdered electrode active substance used for batteries or a powdered large surface material with a large surface area used for an electric double layer capacitor is made to adhere to a current-collecting member.

Fig. 1 shows a process for manufacturing an electrode structure 1 wherein a powdered electrode substance, i.e., a powdered electrode active substance 11, comprising particles of a compound such as  $\text{LiCoO}_2$  is coated with an ion-conducting polymer 12, and is made to adhere to a current-collecting member 13. Likewise, Fig.

2 shows a process for manufacturing the electrode structure 1 wherein a powdered electrode substance, i.e., the powdered electrode active substance 11 having a form such as that of graphite or hard carbon is coated with the ion-conducting polymer 12, and is made to adhere to the current-collecting member 13. A powdered large surface material such as an activated carbon as the powdered electrode substance may be coated with the ion-conducting polymer and is adhered to the current-collecting member to form an electrode structure of the electric double layer capacitor.

Fig. 1 shows the case where the electrical conductivity of the powdered electrode active substance 11 is low. The electrical conductivity in the powdered electrode active substance and between the powdered electrode active substance 11 and current-collecting member 13, is increased and the current-collecting efficiency improved by mixing with a powdered electrically-conducting substance 14.

The powdered electrically-conducting substance 14 may or may not be coated with the ion-conducting polymer.

"Coating" refers to a state of contact wherein ions can easily migrate between the ion-conducting polymer 12 and the powdered electrode substance, i.e., the powdered electrode active substance 11 over their entire surfaces or the powdered large surface material over their entire surfaces. The ion-conducting polymer 12 is coated on the surface of the powdered electrode active substance 11 or the powdered large surface material so that the latter is covered by the ion-conducting polymer 12. The powdered electrode active substance 11 becomes more active the finer the particles of which it is comprised, but its activity can be suppressed to give greater stability by coating with the ion-conducting polymer 12.

If the layer of the coated ion-conducting polymer 12 is thick, conductivity

decreases and current-collecting efficiency is poorer, so it is preferably thin.

The term "powdered" as used in the powdered electrode active substance 11, the powdered electrically-conducting substance 14, or the powdered large surface material refers to a substance having a fine particle state. In some cases, it may refer to a state wherein a large number of substances having a fine particle state are agglomerated.

For the purpose of obtaining a desirable electrode structure 1, the roller is activated to role the electrode structure 1 having an application layer 15 formed by applying the powdered electrode active substance 11 on the current-collecting member 13 in order to strengthen the adhesiveness between the powdered electrode active substance 11 and the current-collecting member 13 and to increase the density of the powdered electrode active substance 11. The rolled electrode structure may be a conventional structure or a structure coated with the ion-conducting polymer 12.

#### (c) Rolling device

The rolling device at least is able to rotate and press and has a structure as shown in Fig. 3(A). In this rolling device, the tape electrode structure body 1, comprising the current collecting member 13 superposing an application layer 15 containing the powdered electrode active member as shown in Fig. 3(B), while under the control by a control device 5, is withdrawn from a unwinding roll 53, pre-treated by a preliminary treatment section 51, rolled by roller 4, post-treated by a subsequent treatment section 52, and wound on a winding roll 54.

When in pre-treating, a tensile force and thickness of the tape electrode structure body 1 may be measured, and the electrode structure body 1 may be cleaned. Alternatively, when in post-treating, a tensile force and thickness of the



tape electrode structure body 1 may be measured, and the electrode structure body 1 may be cleaned.

#### (d) Roller

An example of roller 4 is explained with reference to Fig. 4 and Fig. 5. The roller 4 in Fig. 4 has a pair of work rolls 43, 43 and a pair of backup rolls 42, 42 pressurizing the work rolls 43, 43. In order to generate higher pressure on the work rolls 43, 43, diameter of the backup rolls 42, 42 may be designed to be larger than the small diameter work rolls 43, 43. The backup roll 42 is manufactured by adhering an extra-hard elastic material on the surface of an extra-hard material, which prevent from damaging the surface of the work roll 43 due to the contact with the work roll 43. This extra-hard elastic material may be denatured urethane or polyimide, and the hardness for example may be 80 or more at Rubber Code Shore D Scale of JIS but preferably between 90-98. The thickness of the extra-hard elastic material for example is between 100mm-30mm. Positioning the elastic material on the surface, oil and grease adversely affecting on the electrode structure body 1 may be omitted. The work roll 43 may be a small steel roll, being pushed by two large diameter rolls, thereby applying the large pressure on the electrode structure body 1 and applying a rolling force almost fills the void in the coating layer of the electrode structure body 1.

The drive unit 45, such as a motor driving the rolls and the backup roll housing 42 having an axle receiving section of the lower backup roll 42 are positioned at the upper portion of the roller base 41. The pair of work roll housing 443, 443 of the pair of work rolls 43, 43 and a backup roll housing 442 of the upper backup roll are positioned at the upper portion of the lower backup roll housing 422. A roll axle 471 is rotatably installed at the respective axle receiving section 47.

The respective backup roll housing 442 and the respective work roll housing 443, each has a hole through which a diver 441 is penetrated, and the respective work roll housing 443 and the respective backup roll housing 442, each is placed between the roller base 41 and the upper frame plate 415 to penetrate the diver 441 in order to prevent the deviation of the positions of the rolls 42, 43. A gate-shape frame may be used instead of the diver 441. The respective roll housings 442, 443 is desirably positioned at both ends of the rolls 42, 43, where four divers 441 may be employed to stably be fixed.

As necessary, the unwinding core 411 and the winding core 412 are processed in the roller 4, and the electrode structure body 1 is withdrawn from the unwinding core 411 holding the electrode structure body 1, and the pair of work rolls 43, 43 are used to roll the electrode structure body 1, thereby winding by the winding core 412. In that case, the unwinding core 411 and the winding core 412 are installed on the frame 413 fixed on a stationary member such as a housing 442, and the motor 414 is activated to perform the drive control.

#### (e) Pressuring device

A pressuring device 44 is fixed on the upper frame plate 415, and the upper backup roll housing 442 is pressed downward, so that the upper backup roll 42 presses the upper work roll 43 downward, thereby generating the pressure between the pair of work rolls 43, 43. The pressuring device presses both ends of the upper backup roll housing 442 and evenly presses the work roll 43 downward. Although the pressuring device 44 is position in the upper portion in Fig. 4 and Fig. 5, it may be positioned in the lower portion or positioned in either side if the rolls are arranged horizontally.

#### (f) Driving unit

The driving unit 45 transmits the rotating force to a work roll driving gear 453 via a driving belt 451 and what not. As necessary, the work roll driving gear 453 and a backup roll driving gear 452 may be installed on the respective roll axle 471 of the rolls for the respective roll to rotate simultaneously or a main roll is directly driven while the remaining rolls may be dependently driven by a motor but simultaneously with the rotation of the main roll.

(g) Adjustment plate

Placing an adjustment plate 46 between the respective roll housings prevents a pressure failure between the respective rolls due to the deviation in the preciseness of the pressure and the operation by the pressuring device 44, which assures the preciseness of the thickness of the electrode structure body 1.

For example, the adjustment plate 46 may be a spring and a spacer, which maintains clearances among four rolls when a pressuring device such as a hydraulic cylinder is extended. The adjustment plate positioned between the work rolls 43, 43 adjust a gap appropriately so that the work rolls 43, 43 may not contact each other. The spring pushes the housings 442, 443 and rolls 42, 43 evenly with gaps when the pressuring device such as a cylinder moves backward.

(h) Roller for a pair of large diameter and small diameter work rolls

In the roller as shown in Fig. 5, the pair of work rolls 43, 43, one roll is pressed by the large backup roll 42, and the other backup roll 42 is omitted at the large diameter work roll, thereby reducing the number of rolls required to be three which downsizes the roller 4.

In order to increase the pressing force, a pair of work rolls 43, 43 are divided into a large diameter roll and a small diameter roll. The small diameter work roll 43

is pressed by the large backup roll 42, and the other work roll 43 for the large diameter work roll 43 may be omitted.

(i) Operation of the roller

As shown in Fig. 6, the pressuring device 44 pushes the backup roll 42 down  
 5 from the above, and the electrode structure 1 inserted between the upper work roll 43 and the lower work roll 43 is rolled/processed due to the operation of the pressuring device 44. If the diameter of the backup roll is set to be about 300mm while the diameter of the work roll is to be about 150mm, unit-area maximum pressure rotating the electrode structure 1 becomes twice or 2.5 times larger than  
 10 the pressure in the traditional device, which improves cell effectiveness. Therefore, a finer work roll 43 comparing to the conventional work roll may be utilized and at the same time there is no difference due to a roll bent (bending of the work roll 43, 43), thereby maintaining the uniform gap.

Therefore, the gap between the work rolls 43, 43 may be adjusted so that the  
 15 entire width is to be about  $80\ \mu$  for example by adjusting the adjustment plate 46 of the lower work roll 43. Then, when the electrode structure 1 is rolled by the work roll 43, the electrode structure 1 is inserted as the thickness of the adjustment plates 46, 46 of the upper work roll and the lower work roll may be adjusted making an adjustment for the work roll bending in order to uniform the thickness of the entire  
 20 width of the electrode structure 1. In this case, the backup roll 42 may be designed to be a drum-shape (a roll with a clausung treatment) as shown in Fig. 6(B).

Fig. 7 uses three work rolls, wherein the lower work roll has a larger diameter and is hard to be bent; the upper work roll has a smaller diameter so as to apply higher unit-area pressure on the electrode structure 1; pushing back by the backup  
 25 roll 42 adjusts the amount of bending; and a uniform large pressure may be applied

to the electrode structure 1. In addition, the adjustment of the thickness of the electrode 1 and the adjustment of the spacer may be facilitated.

Under the above-condition, for example, the thickness of the electrode structure 1 prior to the rolling is set to be  $150\ \mu$  and the gap of the work roll while rolling is to be  $80\ \mu$ , it was acknowledged that the thickness of the electrode structure after rolling regains and becomes desirable  $100\ \mu$ .

#### (j) Powdered electrode active substance

The powdered electrode active substance may be a material which permits insertion and separation of ions, or a  $\pi$ -conjugated electrically-conducting polymer material.

There is no particular limitation on the electrode active material used as the positive electrode in a non-aqueous electrolytic battery, but in the case of a rechargeable secondary cell, a chalcogen compound permitting insertion and separation of lithium ions or a complex chalcogen compound containing lithium may for example be used.

Examples of chalcogen compounds are  $\text{FeS}_2$ ,  $\text{TiS}_2$ ,  $\text{MoS}_2$ ,  $\text{V}_2\text{O}_5$ ,  $\text{V}_6\text{O}_{13}$  and  $\text{MnO}_2$ . Examples of chalcogen compounds containing lithium are  $\text{LiCoO}_2$ , lithium complexes represented by  $\text{Li}_x\text{Ni}_y\text{M}_{1-y}\text{O}_2$  (where M is at least one metal element chosen from transition metals or Al, but preferably at least one metal element chosen from Co, Mn, Ti, Cr, V and Al, and  $0.05 \leq x \leq 1.10$ ,  $0.5 \leq y \leq 1.0$ ),  $\text{LiNiO}_2$ ,  $\text{LiMnO}_2$  and  $\text{LiMn}_2\text{O}_4$ . These compounds may be obtained from the oxides, salts or hydroxides of lithium, cobalt, nickel or manganese as starting materials, mixing the starting materials depending on the composition, and firing in an oxygen atmosphere at a temperature in the range of  $600\ ^\circ\text{C}$ - $1000\ ^\circ\text{C}$ .

There is no particular limitation on the electrode active substance used as the

negative electrode in a non-aqueous electrolyte battery, but a material permitting insertion and separation of lithium ions may be used such as lithium metal, lithium alloy (alloys of lithium and aluminum, lead and indium, etc.), and carbon materials.

Examples of  $\pi$ -conjugated conducting polymer materials are polyacetylenes, polyanilines, polypyrroles, polythiophenes, poly- $\rho$  (para)-phenylenes, polycarbazoles, polyacenes and sulfur polymers.

In particular, in non-aqueous electrolyte primary cells, a large battery capacity can be obtained by using lithium metal for the negative electrode.

In non-aqueous electrolyte secondary cells, an excellent cycle life can be obtained by using carbon materials which permit insertion and separation of lithium as the negative electrode. There is no particular limitation on the carbon material, examples being pyrolytic carbon, cokes (pitch coke, needle coke or petroleum coke), graphites, glass carbons, organic polymer compound firing products (products obtained by firing and carbonizing phenolic resins and furan resins at a suitable temperature), carbon fiber and active carbon.

(k) Powdered electrode substance with a large surface area

A powdered electrode substance with a large surface area is a powdered large surface material that may draw many ions on that surface.

A preferable powdered large surface material is a carbon material with a specific surface area of  $500\text{m}^2/\text{g}$  or more, preferably  $1000\text{m}^2/\text{g}$  or more, and more preferably most preferably  $1500\text{m}^2/\text{g}$ - $3000\text{m}^2/\text{g}$  and an average particle diameter of  $30\mu\text{m}$  or less, preferably  $5\text{-}30\mu\text{m}$ . The specific surface area and the average particle diameter outside the above-ranges cause a large electrostatic capacity and make it difficult to obtain an electric double layer capacitor with a lower resistance.

A preferable powdered large surface material especially is an activated carbon resulting from activating the carbon material such as by a steam activating treatment and a fused KOH activating treatment. The activated carbon for example may be palm shell activated carbon, a phenol activated carbon, a petroleum coke type activated carbon, mesofusepitch carbon, and polyacenes. One of the above activated carbon types or combination of two or more activated carbon types may be employed. Phenol activated carbon, a petroleum coke type activated carbon, and a polyacenes are preferable since they provide a larger electrostatic capacity.

(l) Powdered electrically-conducting substance

The powdered electrically-conducting substance increases the electrical conductivity of the electrode structure, there being no particular limitation thereon, but metal powders and carbon powder may be used. As carbon powder, pyrolytic carbon such as carbon black and its graphitization products, artificial and natural scaly graphite powder, and carbon fiber and its graphitization products, are suitable.

Mixtures of these carbon powders may also be used.

(m) Ion-conducting polymer

The ion-conducting polymer is a polymer which can dissolve at least the lithium salts described hereafter at a concentration of at least 0.1M (moles/l), the polymer containing the lithium salt at a concentration of at least 0.1M having an electrical conductivity of  $10^{-8}$  S (siemens)/cm at room temperature. It is to be particularly preferred that the ion-conducting polymer dissolves at least lithium salts to a concentration of 0.8M-1.5M, the resulting polymer solution having an electrical conductivity of  $10^{-3}$  S/cm -  $10^{-5}$  S/cm at room temperature.

The lithium salt is at least one type of lithium salt having  $\text{ClO}_4^-$ ,  $\text{CF}_3\text{SO}_3^-$ ,  $\text{BF}_4^-$ ,  $\text{PF}_6^-$ ,  $\text{AsF}_6^-$ ,  $\text{SbF}_6^-$ ,  $\text{CF}_3\text{CO}_2^-$  or  $(\text{CF}_3\text{SO}_2)_2\text{N}^-$  as anion.

5           The current-collecting member should be a substance which easily passes electricity. Its shape and material are selected according to the electrical components involved, and as an example, it can be formed by fashioning an electrically-conducting substance such as aluminum or copper into a plate, foil or mesh.

(o) Secondary cell

(i) Electric double layer capacitor

15



substance is disposed between the pair of electrode structures.

The method of manufacturing these electrode structures will now be described.

(a) Manufacture of electrode structure

If the powdered electrode active substance is used as the powdered electrode  
5 substance, to manufacture the electrode structure, an extremely thin ion-conducting  
polymer or ion-conducting polymer raw material is coated on the surface of the  
powdered electrode active substance 11. Next, a solvent is added to liquefy the  
mixture into a paste which is applied to the current-collecting member, and dried to  
evaporate the solvent. Alternatively, the solvent can be added from the beginning  
10 to make a paste when the ion-conducting polymer or ion-conducting polymer raw  
material is coated. If the powdered large surface material is used as the powdered  
electrode substance, the same process applied with the powdered electrode active  
substance may be employed to manufacture the electrode structure.

In this process, a minute amount of the ion-conducting polymer or ion-  
15 conducting polymer raw material is used, the particle surfaces of the powdered  
electrode active substance are coated with the ion-conducting polymer, no voids are  
formed, and gaps in the powdered substance are reduced.

To coat the ion-conducting polymer or ion-conducting polymer raw material on  
the powdered electrode active substance, the ion-conducting polymer or ion-  
20 conducting polymer raw material and the powdered electrode active substance are  
press-slid together to obtain a press-slid product.

(b) Press-sliding

Press-sliding is the action of sliding while pressing mixtures 10 of the ion-  
conducting polymer 12 or the raw material of the ion-conducting polymer 12 and the  
25 powdered substance 11 together. An external force is applied to the mixtures so

that they cohere to each other and the particles rotate, and this process is performed repeatedly to obtain a press-sliding product.

#### (c) Press-sliding mixer

The press-sliding mixer is shown, for example, in Fig. 4. The mixture 10 of the ion-conducting polymer 12 or its raw material with the powdered substance 11, or the mixture 10 comprising this mixture and a solvent or the like, is introduced into a container 21, and the main blade 22 is rotated. There is a gap between a base 211 of the container 21 and a bottom surface of the main blade 22. When the main blade 22 is rotated, part of the mixture 10 enters the space between the base 211 of the container 21 and the main blade 22, is subjected to press-sliding, and is kneaded. This process is repeated so that the ion-conducting polymer 12 or its raw material coats the powdered substance 11.

A press-sliding mixer 2 may if necessary be provided with a dispersion blade 23 in the container 21. The dispersion blade 23 is rotated at high speed to disperse the press-slid mixture 10.

#### (d) Container

The container 21 is provided for holding the mixture 10 which is press-slid and stirred. The bottom surface of the container 21 may be such as to permit press-sliding of the mixture 10, and may be slanting as in Fig. 4 or flat as in Fig. 5. For example, if it is slanting, it has a bottom part 2111, and slants upwards from the bottom part 2111 towards the circumference. The center part may be situated in a low position, and have a slope rising towards the circumference. The bottom 211 may be formed in the shape of, for example, a grinding mortar, and the angle of the bottom part 2111 may for example be 120 degrees. The bottom 211 of the container is wear-resistant, and is formed by thermal spraying with tungsten or

carbide using SUS. Plural bottom parts 2111 of this type may also be formed on the bottom surface.

(e) Main blade

The main blade 22 functions together with the bottom surface of the container 21, serving to press-slide and stir the mixture. In the main blade 22, a shaft is attached in a position corresponding to the bottom part 2111 of the container 21 as shown for example in Fig. 9(B), and it curves upwards along the bottom of the container from the bottom part 2111. The main blade 22 may comprise two blades attached from the center part as shown in Fig. 9(B), or it may comprise a larger number of blades, e.g. 10 or more, depending on the amount and type of mixture.

The rotation speed of the main blade, i.e., the number of rotations, is set low, for example to 120 rpm or less, when press-sliding is performed.

The gap between the bottom surface of the container 21 and the base surface of the main blade 22 is set as narrow as is necessary for press-sliding the mixture, for example 15 mm or less.

The surface in the motion direction (press-sliding direction) of the main blade 22 is formed so that a pressing angle  $\theta$  relative to the bottom surface of the container 21 is an acute angle. For example, if the cross-section of the main blade 22 is a reverse trapezoid as shown in Fig. 9(C), the pressing angle is from 3 degrees to 70 degrees. The cross-section of the main blade 22 may also be circular or have a rounded corner as shown in Fig. 9(D). The material of the main blade has wear-resistant properties, and is formed for example by thermal spraying with tungsten or carbide using SUS.

If a surface in a direction opposite to the motion direction (press-sliding direction) of the main blade 22 is formed for example effectively perpendicular to or

at an obtuse angle to the bottom surface, the mixture 10 can be collected around the main shaft 221 by rotating the main shaft 221 in the reverse direction.

(f) Dispersing blade

The dispersion blade 23 is intended to disperse the mixture 10 which has been press-slid by the main blade 22. The dispersion blade 23 is disposed in a position at which the mixture 10 can be dispersed, and it rotates at a high speed such as 1000-4000 rpm. Due to this high speed rotation, the ion-conducting polymer 12 or its raw material coated on the particle surfaces of the powdered substance 11 uniformly disperses through the whole of the powdered substance. The mixture adheres on the area surrounding the main shaft 221 of the main blade 22 when in firm mixing or low clay dispersion and tends to be ununiformed mixing. The dispersion blade 23 is positioned on the main shaft 221 of the main blade 22, rotating the same from the dry mixing stage to prevent the adhesion on the area surrounding the main shaft 221.

(g) Action of press-sliding mixer

A description of the operation using the press-sliding mixer of Fig. 10 and Fig. 11 will now be given. The press-sliding mixer is supported on a supporting platform 24, the container 21 being raised by a handle 241, and is controlled by a control panel 25.

First, the powdered substance 11 (containing an additive) is measured out, and introduced from a powdered substance input port 34. In an automated system, a measuring hopper or the like is installed above the powdered substance input port 34 for storage and measurement, and a valve 341 of the powdered substance input port is automatically opened by an input command. Simultaneously, to eliminate measurement errors from pressure rise in the container due to introduction of the



penetration dispersion of the ion-conducting polymer or its raw material in the powdered substance is promoted. Here, care should be taken when a low boiling-point solvent is added to the ion-conducting polymer or its raw material to promote dispersion in the powdered substance, as the concentration or viscosity of the ion-conducting polymer or its raw material increases and dispersion becomes difficult if suction degassing is continuously performed by a high vacuum blower.

When penetration degassing has reached about 70% after degassing for approximately 1 hour, the dispersion blade is rotated at 2800 rpm to promote dispersion.

#### 10 (h) Application to current-collecting member

The press-slid substance which has been made into a paste is thinly applied to the surface of the current-collecting member. After application, the solvent evaporates and dries to obtain the electrode structure. The device which applies the press-slid substance to the current-collecting member may be a doctor knife applicator. The applied electrode structure is applied to the tape current collecting member as shown so that the application layer at the respective electrode is isolated in Fig. 3(B).

#### (a) Rolling by the rolling device

The electrode structure 1 is set between the work rolls 43 and 43, and stall tension (tension at the stand by state and 20-30% tension at driving) is applied to the electrode structure 1 between the unwinding rollers 53 and 54. When the line is shifted in a yield drive, the entire line is driven in extremely low speed. Then, the upper backup roll housing 442 and the upper backup roll 42 begin to move downward due to the operation of the pressuring device. The upper backup roll 42 and the upper work roll 43 make a line contact, and the upper work roll 43 together

with the upper backup roll 42 rotate and move downward. The upper work roll 43 contacts the upper surface of the electrode structure 1; three rolls contact and roll; if the rolls move downward further by pressing the spring, the lower work roll 42 comes to contact with the surface of the lower backup roll 42. As a result, four rolls  
 5 synchronize and begin to make low r.p.m. operation.

The pressuring device 44 such as a hydraulic cylinder further pressurize the upper backup roll housing 442 with the predetermined pressure, and roll surfaces of four rolls are evenly pressurized.

If the work rolls 43, 43 intensively rotates the electrode structure 1, the  
 10 application layer 15 and the current collecting member 13 are compressed together to be compressed and expanded in the cross direction to be deformed. On the other hand, a layer without the application layer 15 is not compressed, and therefore no compression occurs. Accordingly, the boundary section between the application layer 15 and the layer without the application layer 15 brake and the strength is  
 15 decreased. Limit of this tensile force is considered to be about twice as the tensile force by the unwinding roller 54.

If the tape electrode structure 1 is rolled, the pressing force on the work rolls 43, 43 is increased, and the best rolling with high compressibility prior to the breakage of the electrode structure 1.

20 The conventional roller is comprised of two work roles without a backup roll, and the pressing force may not be increased due to the distortion of the work roll. On the other hand, the roller 4 in this invention is capable of rolling the electrode structure 1 until it breaks and to obtain the electrode structure 1 with high compressibility before the breakage, thereby obtaining high electrode density and  
 25 low electrode impedance.

By rolling the electrode structure 1 using the roller 4 of this invention, the density of the electrode structure increases. The electrode density increase means an increase in the area of contact between the powdered electrode active substances in the electrode structure and between the powdered electrode active substance and the powdered electrode conducting substance in the electrode structure. As a result, impedance of the electrode decreases. At the same time, the area of contact between the current collecting member and the powdered electrode active substance and the powdered electrode conducting substance may be set larger. Impedance of the electrode itself decreases to become a desirable electrode. Yet, the electrode density increases, and the amount relative to unit-area surface of the powdered electrode active substance and the powdered electrode conducting substance in the electrode structure 1 increases. As a result, if the electrode structure 1 is used for a cell, the area-unit energy density increases, thereby the entire cell becomes smaller.

#### **Description of the Preferred Embodiments**

Hereafter, an embodiment of a lithium ion secondary cell will be described.

##### **(a) Example of manufacture of positive electrode structure (Example 1)**

9.1 weight parts of  $\text{LiCoO}_2$  of average particle size  $5\ \mu\text{m}$  which is a powdered electrode active substance, and 0.6 weight parts of graphite powder of average particle size  $4\ \mu\text{m}$  which is a powdered electrically-conducting substance, were introduced into a press-sliding mixer, and press-slid for 20 minutes. Next, 0.546 weight parts of an ion-conducting polymer raw material (A1) and 3.5 weight parts of acetonitrile were added. The ion-conducting polymer raw material (A1) was a mixture, and its composition and mixing ratio are shown in Table 1.

[Table 1]



The press-slid substance to which the ion-conducting polymer raw material (A1) was added, was press-slid in the press-sliding mixer for 5 hours. The press-slid substance was paste-like. 0.254 weight parts of polymeric MDI, MR-200 (NPU Co.) was added to the press-slid substance, and the mixture stirred for 5 minutes in the press-sliding mixer. The press-slid substance was removed, transferred to aluminum foil of thickness  $20\ \mu\text{m}$ , and spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap. The resulting product was left at room temperature for 15 minutes, and then heated at  $80^\circ\text{C}$  for 1 hour. The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at  $80^\circ\text{C}$  for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness  $20\ \mu\text{m}$  gap, and the entire thickness was  $170\ \mu\text{m}$ . This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was  $120\ \mu\text{m}$  gap.

(b) Example of manufacture of positive electrode structure (Example 2)

9.0 weight parts of  $\text{LiCoO}_2$  of average particle size  $5\ \mu\text{m}$  which is a powdered electrode active substance, and 0.6 weight parts of ketjenblack and 0.2 weight parts of graphite powder of average particle size  $4\ \mu\text{m}$  which are powdered electrically-conducting substances, were introduced into a press-sliding mixer (capacity 300cc), and press-slid for 20 minutes. Next, 1.172 weight parts of an ion-conducting polymer raw material (A1) and 3.5 weight parts of acetonitrile were added. These mixtures were press-slid for 5 hours in the press-sliding mixer. The press-slid substance was paste-like. 0.548 weight parts of polymeric MDI, MR-200 (NPU Co.),

was added to the press-slid substance, and the mixture press-slid for 5 minutes.

The press-slid substance was removed, transferred to aluminum foil of thickness 20  $\mu$  m, and spread by a doctor knife applicator of 100  $\mu$  m gap. The resulting product was left at room temperature for 15 minutes, and heated at 80°C for 1 hour. The

5 electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of 100  $\mu$  m gap similarly.

The resulting product was left at room temperature for 15 minutes, and then heated at 80°C for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m.

10 This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was 120 $\mu$ m gap.

#### (c) Example of manufacture of positive electrode structure (Example 3)

9.1 weight parts of LiCoO<sub>2</sub> of average particle size 5  $\mu$  m which is a powdered electrode active substance, 0.341 weight parts of an ion-conducting polymer raw material (A1) and 3.0 weight parts of acetonitrile were introduced into a press-sliding mixer (capacity 300cc), and the mixture press-slid for 7 hours. The press-slid substance was paste-like. Next, 0.159 weight parts of polymeric MDI, MR-200 (NPU Co.) was added, and the mixture press-slid for 5 minutes. The press-slid

substance was removed, transferred to aluminum foil of thickness 20  $\mu$  m, and

20 spread by a doctor knife applicator of 100  $\mu$  m gap. The resulting product was left at room temperature for 15 minutes, and heated at 80°C for 1 hour. The thickness of the electrode obtained was 80  $\mu$  m. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m. This electrode of the invention is rolled by three rollers at

room temperature. The thickness of the electrode after rolling was 120 $\mu$ m gap.

(d) Example of manufacture of positive electrode structure (Example 4)

9.1 weight parts of  $\text{LiCoO}_2$  of average particle size 5  $\mu$  m which is a powdered electrode active substance, and 0.6 weight parts of graphite powder of average particle size 4  $\mu$  m which is a powdered electrically-conducting substance, were introduced into a press-sliding mixer device (capacity 300cc), and press-slid for 20 minutes. Next, 2.0 weight parts of an ion-conducting polymer raw material (A2) and 3.0 weight parts of acetonitrile were added. The ion-conducting polymer raw material (A2) was a mixture, and its composition and mixing ratio are shown in Table

2. In the Table 2, the same result may be obtained by the use of polyethyleneglycoldimethacrylate (536 molecular weight) instead of trimethylolpropanetrimethacrylate.

[Table 2]

The press-slid substance to which the ion-conducting polymer raw material (A2) was added, was press-slid in the press-sliding mixer (capacity 300cc) for 5 hours. The press-slid substance was paste-like. A solution of 0.01 weight parts of 2,2'-azobis (2,4-dimethylvaleronitrile) and 0.5 weight parts of a (1/1) vol liquid electrolyte of ethylene carbonate (EC)/diethylene carbonate (DEC) was added to the press-slid substance, and the mixture further press-slid for 5 minutes. The press-slid

substance was removed, transferred to aluminum foil of thickness 20  $\mu$  m, and spread by a doctor knife applicator of 100  $\mu$  m gap. The resulting product was left at room temperature for 15 minutes, and heated at 80  $^{\circ}\text{C}$  for 3 hours. The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of 100  $\mu$  m gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at

80°C for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m.

This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was 120 $\mu$ m gap.

5 (e) Example of manufacture of negative electrode structure (Example 5)

9.1 weight parts of graphite powder of average particle size 5  $\mu$  m which is a powdered electrode active substance, 0.341 weight parts of an ion-conducting polymer raw material (A1) and 3.0 weight parts of acetonitrile were introduced into a press-sliding mixer (capacity 300cc), and the mixture press-slid for 7 hours. The  
 10 press-slid substance was paste-like. Next, 0.159 weight parts of polymeric MDI, MR-200 (NPU Co.) was added, and the mixture press-slid for 5 minutes. The press-slid substance was removed, transferred to copper foil of thickness 20  $\mu$  m, and spread by a doctor knife applicator of 100  $\mu$  m gap. The resulting product was left at room temperature for 15 minutes, and heated at 80°C for 1 hour. The  
 15 electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of 100  $\mu$  m gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at 80°C for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m.  
 20 This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was 120 $\mu$ m gap.

(f) Example of manufacture of positive electrode structure (Example 6)

9.1 weight parts of graphite powder of average particle size 5  $\mu$  m which is a powdered electrode active substance, 0.2 weight parts of an ion-conducting polymer

raw material (A2) and 3.0 weight parts of acetonitrile were introduced into a press-sliding mixer (capacity 300cc), and press-slid for 5 hours. The press-slid substance was paste-like. A solution of 0.01 weight parts of 2,2'-azobis (2,4-dimethylvaleronitrile) and 0.5 weight parts of a liquid electrolyte of ethylene carbonate (EC)/diethylene carbonate (DEC) in a volume ratio of 1:1 was added to the press-slid substance, and the mixture further press-slid for 5 minutes. The press-slid substance was removed, transferred to copper foil of thickness  $20\ \mu\text{m}$ , and spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap. The resulting product was left at room temperature for 15 minutes, and heated at  $80^\circ\text{C}$  for 3 hours. The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at  $80^\circ\text{C}$  for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness  $20\ \mu\text{m}$  gap, and the entire thickness was  $170\ \mu\text{m}$ . This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was  $120\ \mu\text{m}$  gap.

(g) Comparative example of positive electrode structure (Comparative Example 7)

11.5 weight parts of n-methylpyrrolidine containing, in solution, 0.5 weight parts of polyvinylidene fluoride (PVDF) which has no ion-conducting property as a polymer binder, was mixed with 9.0 weight parts of  $\text{LiCoO}_2$  of average particle size  $5\ \mu\text{m}$  which is a powdered electrode active substance, and 0.8 weight parts of ketjenblack and 0.2 weight parts of graphite powder of average particle size  $4\ \mu\text{m}$  which are powdered electrically-conducting substances, in an ordinary blade mixer. After mixing for 8 hours, the mixture was removed, transferred to copper foil of thickness

20  $\mu$  m, and spread by a doctor knife applicator of 100  $\mu$  m gap. The resulting product was left at room temperature for 15 minutes, and heated at 80°C for 3 hours.

The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of 100  $\mu$  m gap

5 similarly. The resulting product was left at room temperature for 15 minutes, and then heated at 80°C for 1 hour. The electrode obtained is the electrode layer

applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m. This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was 120 $\mu$ m gap.

10 (h) Comparative example of positive electrode structure (Comparative Example 8)

25.5 weight parts of n-methylpyrrolidine containing, in solution, 0.5 weight parts of polyvinylidene fluoride (PVDF) which has no ion-conducting property as a polymer binder, was mixed with 9.5 weight parts of graphite powder of average particle size 4  $\mu$  m which is a powdered electrically-conducting substance, in an ordinary blade

15 mixer. After mixing for 8 hours, the mixture was removed, transferred to copper foil of thickness 20  $\mu$  m, and spread by a doctor knife applicator of 100  $\mu$  m gap. The resulting product was left at room temperature for 15 minutes, and heated at 80°C

for 3 hours. The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of 100  $\mu$

20 m gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at 80°C for 1 hour. The electrode obtained is the electrode layer

applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m. This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was 120 $\mu$ m gap.

(i) Comparative example of positive electrode structure (Comparative Example 1)

11.5 weight parts of n-methylpyrrolidine containing, in solution, 0.5 weight parts of polyvinylidene fluoride (PVDF) which has no ion-conducting property as a polymer binder, was mixed with 9.0 weight parts of  $\text{LiCoO}_2$  of average particle size  $5\ \mu\text{m}$  which is a powdered electrode active substance, and 0.8 weight parts of ketjenblack and 0.2 weight parts of graphite powder of average particle size  $4\ \mu\text{m}$  which are powdered electrically-conducting substances, in an ordinary blade mixer. After mixing for 8 hours, the mixture was removed, transferred to copper foil of thickness  $20\ \mu\text{m}$ , and spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap. The resulting product was left at room temperature for 15 minutes, and heated at  $80^\circ\text{C}$  for 3 hours.

The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at  $80^\circ\text{C}$  for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness  $20\ \mu\text{m}$  gap, and the entire thickness was  $170\ \mu\text{m}$ . This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was  $140\ \mu\text{m}$  gap.

(j) Comparative example of positive electrode structure (Comparative Example 8)

25.5 weight parts of n-methylpyrrolidine containing, in solution, 0.5 weight parts of polyvinylidene fluoride (PVDF) which has no ion-conducting property as a polymer binder, was mixed with 9.5 weight parts of graphite powder of average particle size  $4\ \mu\text{m}$  which is a powdered electrically-conducting substance, in an ordinary blade mixer. After mixing for 8 hours, the mixture was removed, transferred to copper foil of thickness  $20\ \mu\text{m}$ , and spread by a doctor knife applicator of  $100\ \mu\text{m}$  gap. The

resulting product was left at room temperature for 15 minutes, and heated at 80°C for 3 hours. The electrode with a single applied surface obtained is turned upside down to be fixed on a flat surface and is spread by a doctor knife applicator of 100  $\mu$  m gap similarly. The resulting product was left at room temperature for 15 minutes, and then heated at 80°C for 1 hour. The electrode obtained is the electrode layer applied on both side of aluminum foil of thickness 20  $\mu$  m gap, and the entire thickness was 170  $\mu$  m. This electrode of the invention is rolled by three rollers at room temperature. The thickness of the electrode after rolling was 140 $\mu$ m gap.

#### (k) Charging/discharging tests

A test lithium ion secondary cell was manufactured using the positive electrode structures manufactured in the examples and comparative examples. The positive electrode and negative electrode were both cut out to have an electrode surface area of 4cm<sup>2</sup>. Completely solid polymer (all polymer), polymer gel electrolyte (polymer gel), liquid electrolyte (liquid) and a separator were sandwiched between the positive electrode and negative electrode to manufacture the test cell. The concentration of lithium salt (supporting electrolyte salt) in the respective electrolytes was arranged to be 1M. This cell was charged at 0.3mA per 1cm<sup>2</sup> of electrolyte surface area to 4.1V, and after allowing to stand for 15 minutes, it was discharged at 0.3mA/cm<sup>2</sup> to 2.7V. Combinations for which two of these charging/discharging cycles were successfully performed, were considered to be combinations for which charging/discharging is possible, and are shown in Table 3. Table 4 shows the contents of the electrolytes listed in Table 3.

[Table 3]

[Table 4]

In the charging/discharging tests of the examples and comparative examples,



test cells using the positive electrode and negative electrode of this invention could be successfully charged and discharged. However, test cells using the electrodes of the comparative examples and a solid or gel electrolyte could not be charged/discharged, although test cells using liquid electrolytes could be charged/discharged.

#### Industrial Field of Application

This invention has the following advantages:

The roller of this invention makes it possible to sufficiently increase the density of application layer by rolling an electrode structure.

This invention further makes it possible to obtain an effective electrode structure.

## TABLES

[Table 1]

Ion-conducting polymer raw material (A1)

Substance	Mixing ratio (weight parts)
Trifunctional (propylene glycol/ethylene glycol) random copolymer, SANNIX FA-103 (PO/EO=2/8, Mw=3,282, Sanyo Chemical Industries, Ltd.)	8.36
Trifunctional polyol, 1,4-butadiol	0.34
Ethylene cyanohydrin	1.27
Reaction catalyst NC-IM (Sankyo Air Products K.K.)	0.03
Total	10

5 [Table 2]

Ion-conducting polymer raw material (A2)

Substance	Mixing ratio (weight parts)
Cyanoethylated/dihydroxypropylated polyvinyl alcohol	0.625
Methoxypolyethyleneglycol methacrylate (mol. wt. 468)	3.125
Trimethylolpropanetri methacrylate	6.25
Total	10

[Table 3]

Charging/discharging test results

No	Positive electrode	Negative electrode	Electrolyt e	Charging/discharging test result
1	Example 1	Example 5	AP1	Possible
2	Example 1	Example 5	AP2	Possible
3	Example 1	Example 5	AP3	Possible
4	Example 1	Example 5	AP4	Possible
5	Example 1	Example 5	PG1	Possible
6	Example 1	Example 5	PG2	Possible
7	Example 1	Example 5	L1	Possible
8	Example 2	Example 5	AP3	Possible
9	Example 2	Example 6	PG2	Possible
10	Example 4	Example 5	AP3	Possible
11	Example 4	Example 6	PG2	Possible
12	Example 7	Example 9	L1	Possible
13	Comparative example 1	Comparative example 2	AP1	Charging/discharging impossible
14	Comparative example 1	Comparative example 2	AP2	Charging/discharging impossible
15	Comparative example 1	Comparative example 2	AP3	Charging/discharging impossible
16	Comparative example 1	Comparative example 2	AP4	Charging/discharging impossible

17	Comparative example 1	Comparative example 2	L1	Possible
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[Table 4]

Electrolyte used in test

Symbol	Type	Composition	Thickness
AP1	All polymer	Cyanoethylated/dihydroxypropylated cellulose (e.g. Japanese Patent Application Laid-Open No. 8-225626)	100 $\mu\text{m}$
AP2	All polymer	Cyanoethylated/dihydroxypropylated cellulose and methacryl polymer 3D cross-linked structure (e.g. Japanese Patent Application Laid-Open No. 8-225626)	100 $\mu\text{m}$
AP3	All polymer	High viscosity polyurethane electrolyte (e.g. Japanese Patent Application No. 11-78085)	100 $\mu\text{m}$
AP4	All polymer	Cyanoethylated/dihydroxypropylated polyvinyl alcohol (e.g. Japanese Patent Application No. 11-78086)	100 $\mu\text{m}$
PG1	Polymer gel	Cyanoethylated/dihydroxypropylated polyvinyl alcohol and methacryl polymer 3D cross-linked structure containing 50% ethylene carbonate (EC)/diethylene carbonate (DEC) = (1/1) vol liquid electrolyte	100 $\mu\text{m}$

		(e.g. Japanese Patent Application No. 11-78087)	
PG2	Polymer gel	High viscosity polyurethane electrolyte containing 50% ethylene carbonate (EC)/diethylene carbonate (DEC) = (1/1) vol liquid electrolyte (e.g. Japanese Patent Application No. 11-78085)	100 $\mu\text{m}$
L1	Liquid	Impregnation of ethylene carbonate (EC)/diethylene carbonate (DEC) = (1/1) vol solution in polyethylene separator	23 $\mu\text{m}$

What is claimed is:

1. A roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising a pair of work rolls sandwiching and rolling the electrode structure,

5 at least a pair of backup rolls generating a pressing force onto the respective work roll,

a pressuring device pressing at least one of said backup roll toward the work roll side, and

a drive unit rotating said work rolls.

10 2. The roller processing/rolling an electrode structure as is claimed in Claim 1, wherein

the diameter of said backup roll is larger than the diameter of said work roll.

3. The roller processing/rolling an electrode structure as is claimed in Claim 1, wherein

15 said backup roll has an elastic material coated on its surface.

4. The roller processing/rolling an electrode structure as is claimed in Claim 1, which further comprises

a work roll housing having an axle receive of said work roll inside and

a housing having an axle receive of the respective roll inside, wherein

20 a spacer is positioned between the respective housings.

5. A roller processing/rolling an electrode structure in which a powdered electrode active substance is adhered to the current collecting member, comprising a large diameter work roll and a small diameter other work roll having an electrode structure inbetween,

25 a backup roll generating a pressing force onto the surface of the small diameter

work roll, and

a pressure device pressing the backup roll toward the small diameter work roll side, and a drive unit rotating the work roll.

6. The roller processing/rolling an electrode structure as is claimed in Claim 5,  
5 wherein

the diameter of said backup roll is larger than the diameter of said work roll.

7. The roller processing/rolling an electrode structure as is claimed in Claim 5,  
wherein

said backup roll has an elastic material coated on its surface.

10 8. The roller processing/rolling an electrode structure as is claimed in Claim 5,  
which further comprises

a work roll housing having an axle receive of said work roll inside and

a housing having an axle receive of the respective roll inside, wherein

a spacer is positioned between the respective housings.

15 9. An electrode structure for a secondary cell in which ions move between the  
electrodes, wherein

a powdered electrode active substance coated by an ion-conducting polymer is  
adhered to a current collecting member, and a roller processes/rolls said powdered  
electrode conductive substance on the current collecting member.

20 10. The electrode structure as is claimed in Claim 9, wherein

said powdered electrode conductive substance is mixed with said powdered  
electrode active substance.

11. An electrode structure of an electric double layer capacitor in which ions  
25 move between electrodes, wherein



a powdered large surface material coated with an ion-conducting polymer is adhered to a current collecting material, and

said powdered large surface material is rolled on said current collecting member by a roller.

5 12. The electrode structure as is claimed in Claim 11, wherein

said powdered large surface material is mixed with said powdered electrode conductive substance.

## ABSTRACT

It is an object of this invention to provide a roller which is effective and suitable for rolling the electrode structure 1 coated with an elastic material on its surface and is to provide a processed electrode structure, the roller comprises a pair of work rolls 43, 43 sandwiching the electrode structure for rolling, at least a pair of backup rolls 42, 42 pressurizing the respective work roll, a pressuring device 44 pressurizing at least one of the backup rolls toward the work roll side, and a drive unit 45 rotating the work roll 43.

Fig. 1

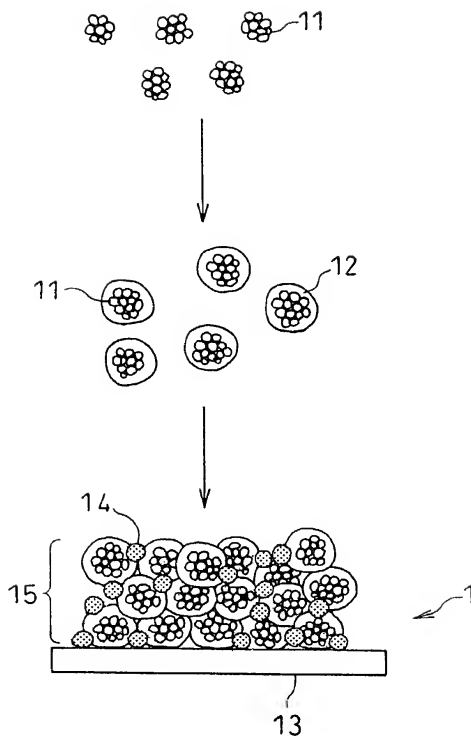


Fig. 2

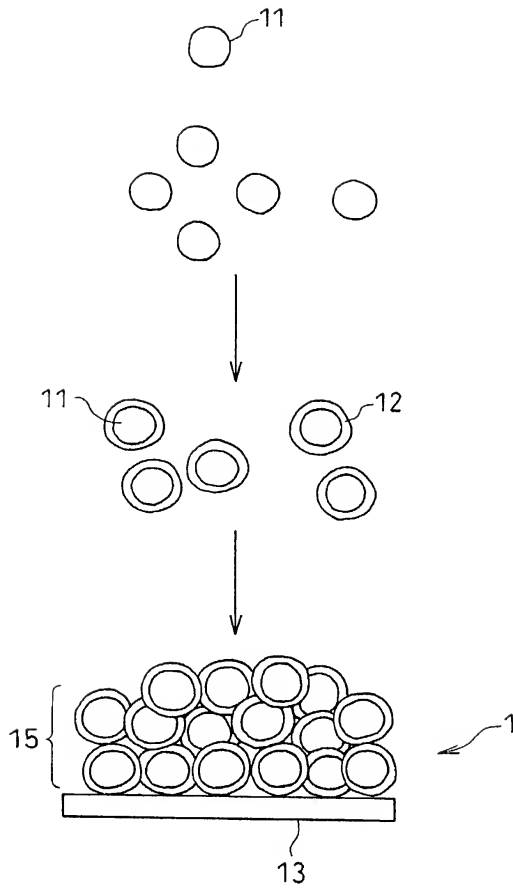
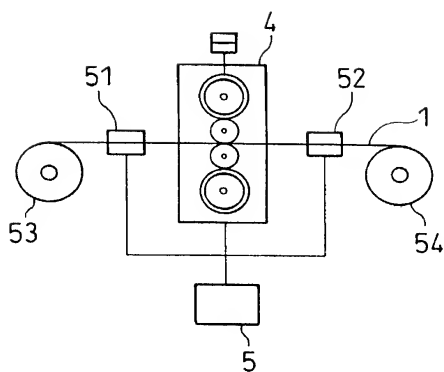


Fig. 3

(A)



(B)

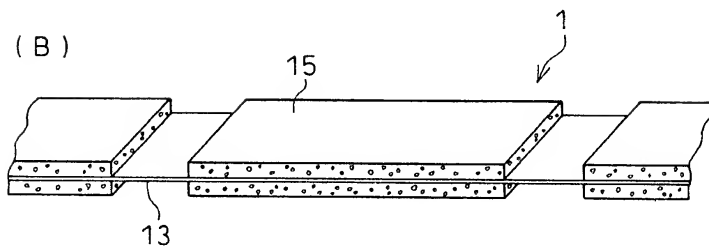




Fig. 5

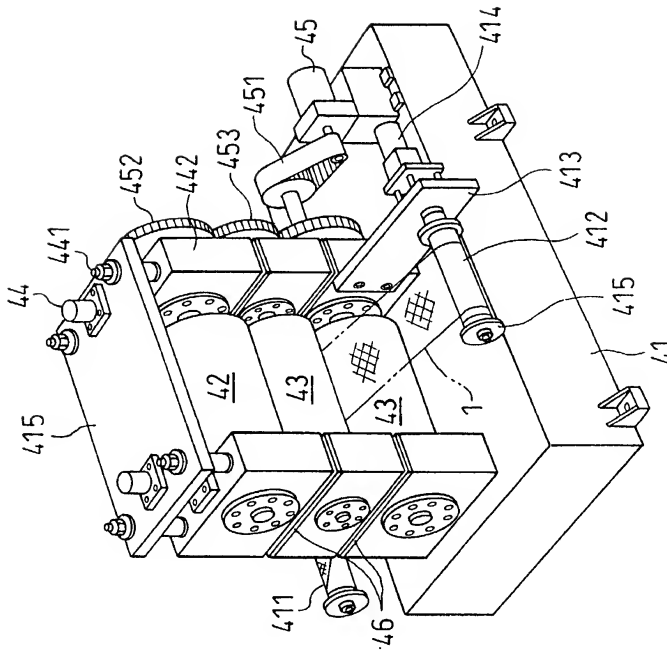


Fig. 6

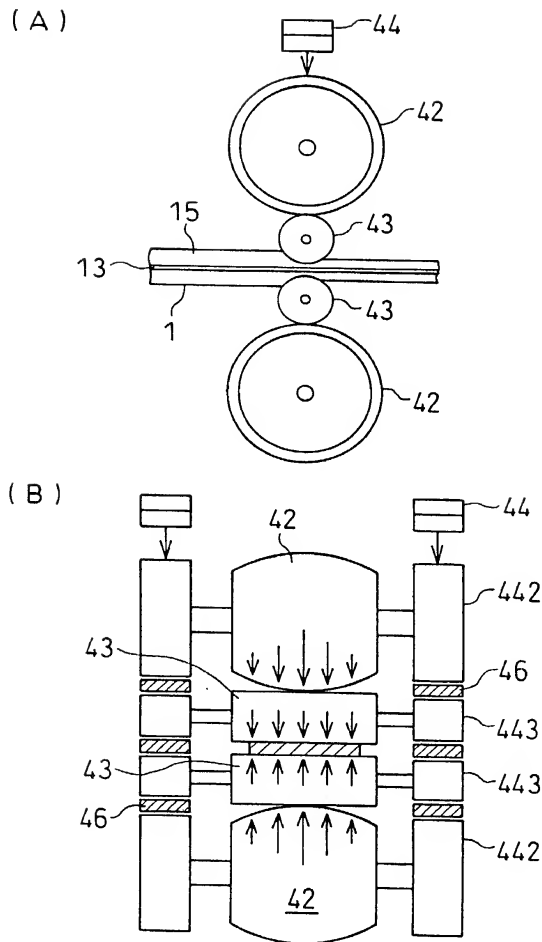




Fig. 7

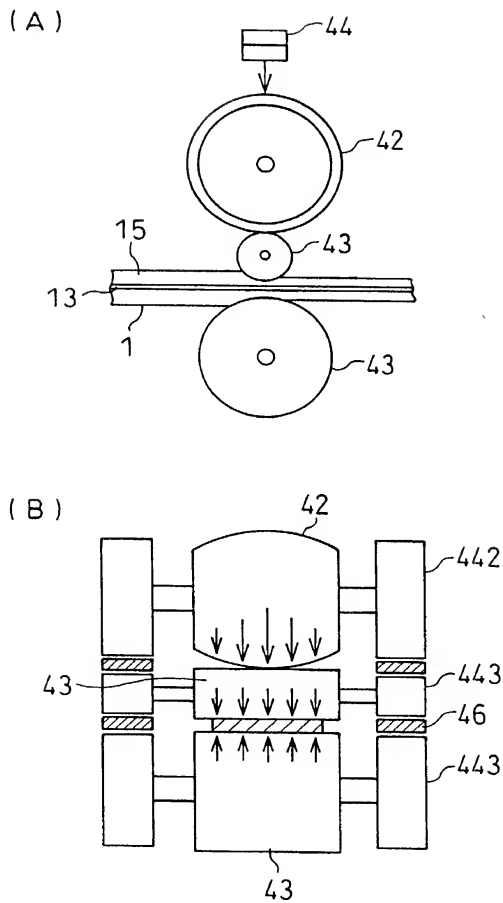


Fig. 8

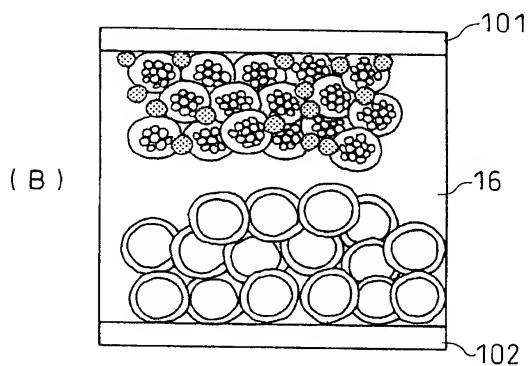
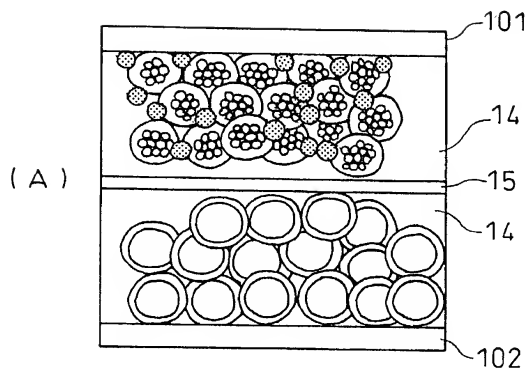


Fig. 9

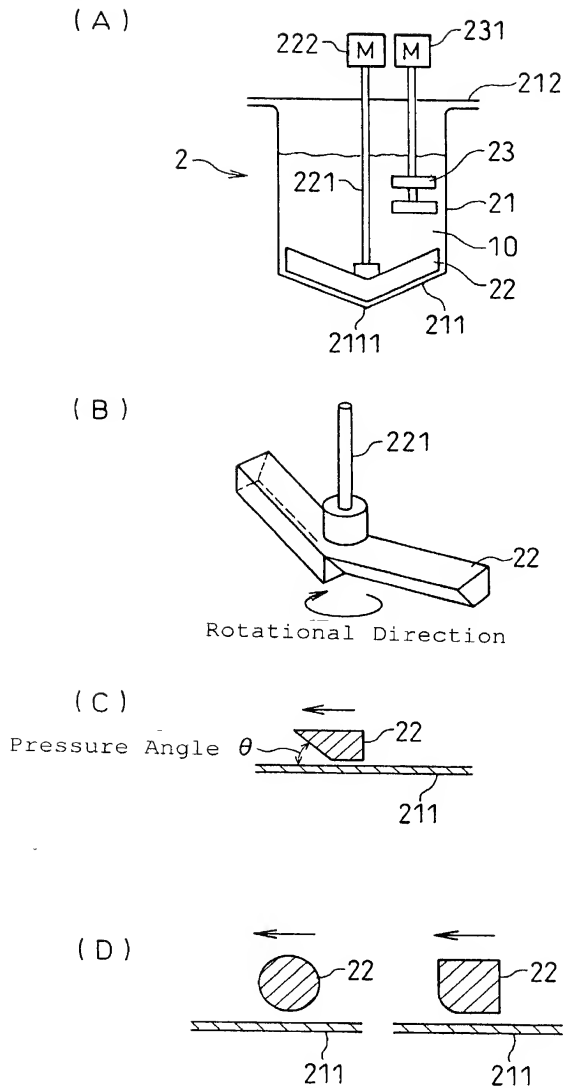


Fig. 10

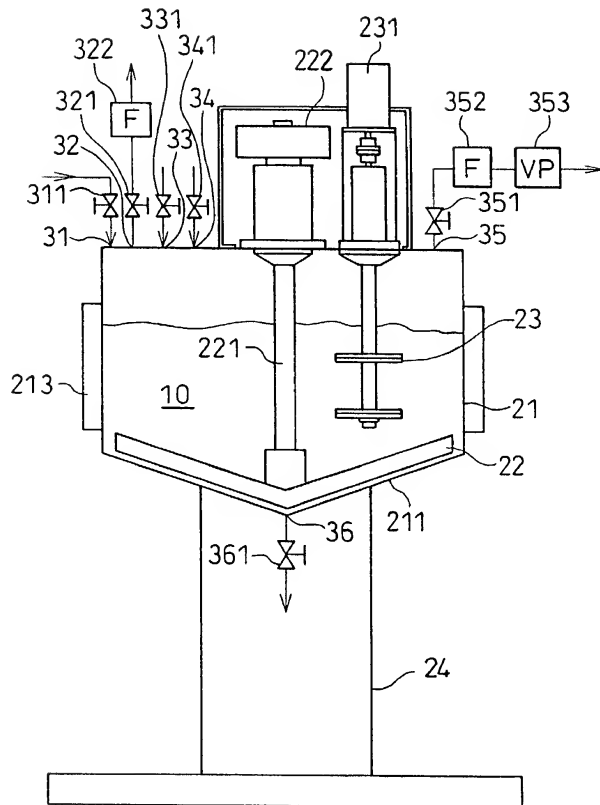


Fig. 11

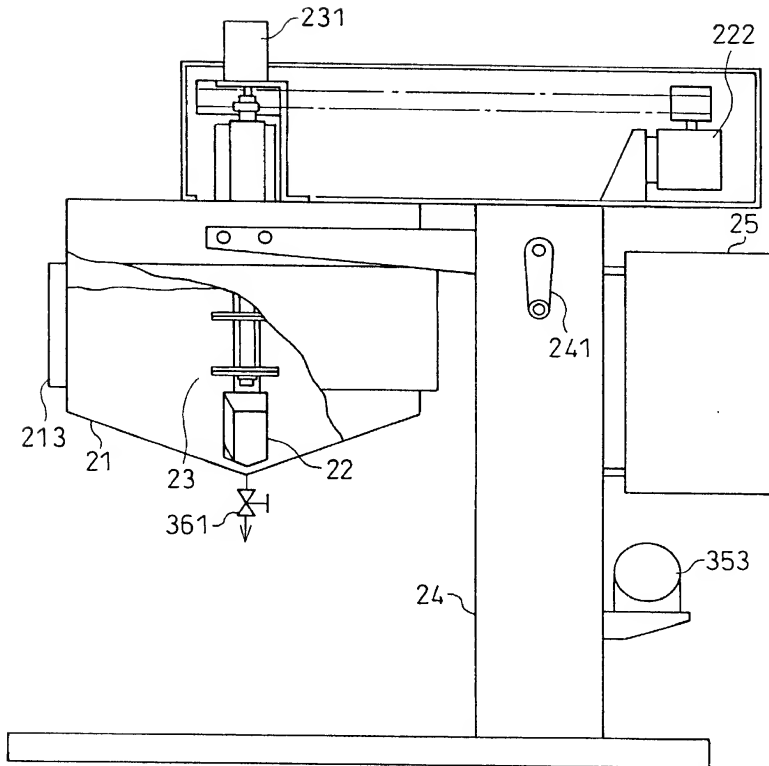


Fig. 12

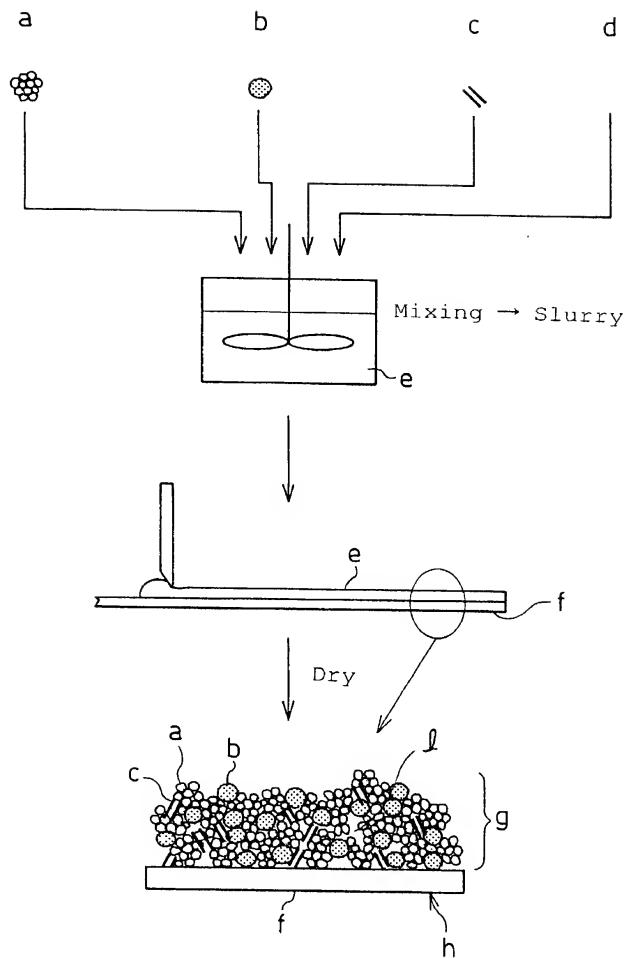




Fig. 14

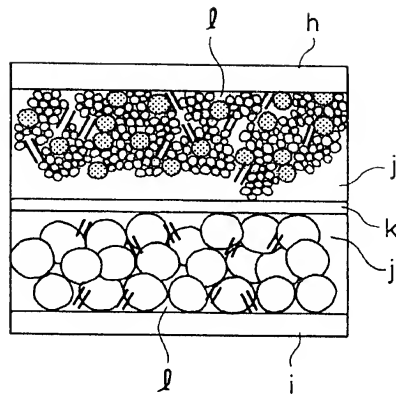
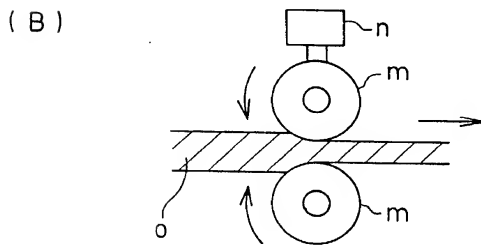
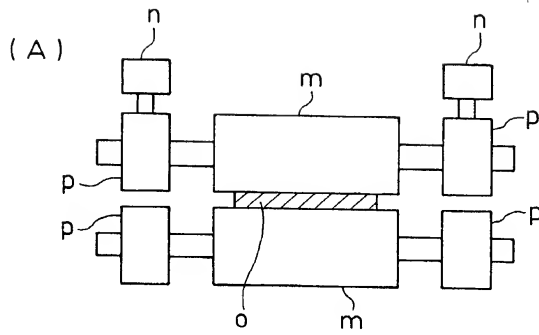




Fig. 15



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My residence, post office address and citizenship are as stated next to my name

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I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

ROLLER FOR ROLLING ELECTRODE

STRUCTURE AND THE ELECTRODE

STRUCTURE

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PCT/JP00/06466 and was amended on  
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Prior Foreign Application(s)

外国での先行出願

Priority Not Claimed

優先権主張なし

11-269124

Japan

22/09/1999

(Number)  
(番号)

(Country)  
(国名)

(Day/Month/Year Filed)  
(出願日/月/年)

☐

(Number)  
(番号)

(Country)  
(国名)

(Day/Month/Year Filed)  
(出願日/月/年)

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(Application No.)  
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PCT/JP00/06466

Sept. 21, 2000

Pending

(Application No.)  
(出願番号)

(Filing Date)  
(出願日)

(Status: Patented, Pending, Abandoned)  
(現況: 特許許可、係属中、放棄)

(Application No.)  
(出願番号)

(Filing Date)  
(出願日)

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(現況: 特許許可、係属中、放棄)

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